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E-BEAM HCl LASER

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United Aircraft Research Laboratories

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E-BEAM HCl LASER

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For the Period May 1, 1973 - November 30, 1973

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13 ABSTRACT

The feasibility of producing stimulated emission in the 3 - 5 wavelength band by exciting hydrogen chloride vibrational levels by electron impact is being investigated theoretically and experimentally. Theoretical predictions of electrically pulsed HCl laser performance indicate that gain coefficient in excess of 1%/cm and optical efficiencies in the 20-30 percent range should be realizable in the present experiment. The pulsed HCl laser experiments in progress are described.

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E-BEAM HCl LASER

I. SUMMARY

Of the infrared transmission windows existing in the sea-level atmosphere which are of interest for Navy and DoD applications, only the 3.5 - 4.0 band lacks a corresponding efficient electric-discharge laser source. The research program described herein describes progress under an ONR/ARPA-sponsored experimental and analytical investigation of a low-pressure gaseous hydrogen chloride (HCl) medium excited by an electron-beam controlled, pulsed electric discharge, in order to develop multiline laser emission on low-lying vibration-rotation transitions in HCl within this desired wavelength band. Consideration of reported swarm experiments in HCl suggests that electron pumping of the vibrational modes of this molecule, might be quite favorable at judiciously chosen discharge conditions.

Theoretical predictions of electrically pulsed HCl laser performance have been performed which indicate that this system should be characterized by optical gains in excess of 1%/cm and optical efficiencies in the 20-30% range. The predictions are derived from the solution to the time-dependent vibrational rate equations governing the rates of change of the HCl vibrational state populations in a 0.1 - 1.0 eV plasma, Provision is made in this analysis for inelastic electron impact excitation, vibration-to-vibration and vibration-to-translation energy exchange, and spontaneous and stimulated emission. Furthermore, consideration of the electron and heavy particle kinetics of the deuterium-hydrogen chloride system suggests that an alternative excitation route may be available, the efficient vibrational energy transfer from D_2 to the HCl emitters, analogous to the well-studied, efficient CO_2/N_2 gas laser system.

During this reporting period, difficulties encountered in obtaining reliable operation of a commerically supplied high-voltage d.c. pulse power supply to energize the high-current electron gun precluded experimental demonstration of this HCl laser. Continuation of attempts to demonstrate laser action in HCl pumped purely by electric discharge is planned.

II. INTRODUCTION

Present Contract Research

Since mid-April 1972 the United Aircraft Research Laboratories (UARL) have been engaged in a research program sponsored by ONR/ARPA, under Navy Contract N00014-72-C-0450 (Ref. 1) to explore the possibility of generating stimulated emission from the vibrational-rotational transitions of hydrogen chloride excited by electron-beam controlled electric discharge. Theoretical studies carried out before the Contract award had indicated the possibility of laser action in hydrogen chloride pumped by electron impact; however, these calculations also indicated that careful control of experimental parameters would be necessary for a successful demonstration. For these reasons, an experimental investigation of HCl excited by a pulsed discharge, sustained by a pulsed electron beam, was proposed and has been pursued in the subject research program. Subsequent investigations (Ref. 2 and 3) have indicated the likelihood of strong vibrational excitation by electron impact in HCl. Results obtained at other laboratories (Ref. 4 and 5) have shown vibrational excitation and development of stimulated emission from the analogous hydrogen halides: HF and DF. These results are summarized within Section III.

Due to unforeseen difficulties encountered in developing the state-of-the-art components within the electron beam generation system necessary in order to provide conditions conducive to the generation of stimulated emission from hydrogen chloride, a demonstration has not as yet been attempted. The problems which have presented themselves, seem at this point to be surmountable. Progress in addressing these problems is discussed herein. The results of extensive computations describing the HCl electric discharge media are also discussed. These calculations appear to be quite encouraging.

III. TECHNICAL RESULTS

Theoretical Investigations

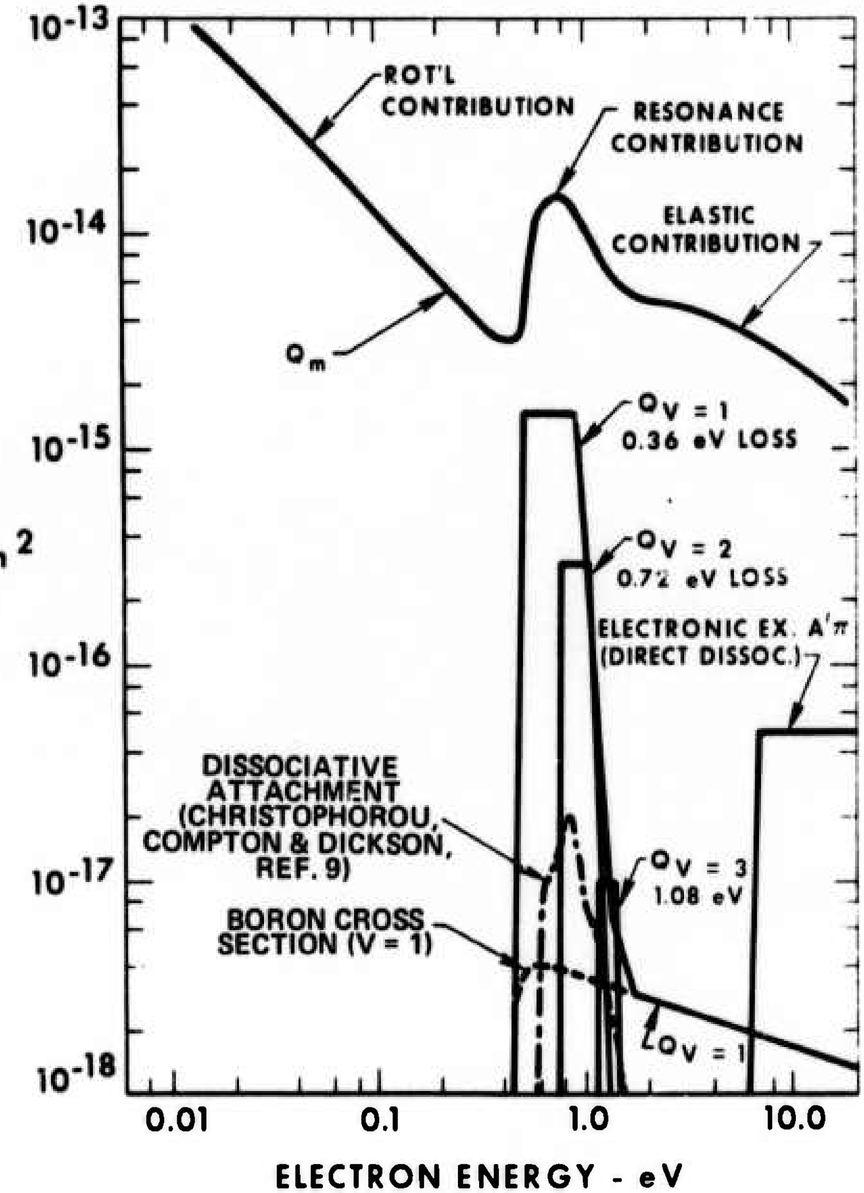
Electron-Molecule Excitation Rates

Analytical prediction of electrically-excited HCl laser performance requires a reasonable estimate of the cross-sections for vibrational excitation of HCl via electron impact. Therefore, considerable analytical effort has been devoted to the theoretical interpretation of the experimental transport data appearing in the literature descriptive of electron-HCl collisional interactions (Ref. 1). These analyses were conducted to reveal as much as possible regarding the electron-HCl vibrational excitation process. The cross-sections inferred from these analyses have been corroborated to a considerable extent by the results of electron transmission experiments conducted by Professor G. Schulz's laboratory at Yale University (Ref. 3). The transport data analysis and the transmission experiments both indicate a peak value of about 1.5×10^{-15} cm^2 for the cross section for excitation of the first HCl vibrational state.

The early electron-HCl transport data of Bailey and Duncanson (Ref. 6) indicate unusually large electron energy losses in HCl for characteristic electron energy values below 1.0 eV. Reasonable estimates of the electron drift velocity v_d and characteristic electron energy ϵ_K can be deduced from this data as a function of the Townsend parameter. Using these data in conjunction with other techniques (Ref. 7), the Frost and Phelps technique (Ref. 8) for the analysis of transport coefficients was used to determine a self-consistent set of electron-HCl cross sections.

Analysis of the Bailey and Duncanson transport data requires numerical solution of the electron Boltzmann equation using a complete trial set of HCl cross sections. The computed electron energy distribution function is then used to calculate the electron drift velocity and characteristic energy which are compared with experimental data. Adjustments are made to the initial cross section set in a trial and error fashion until the calculated and experimental transport data are in reasonable agreement. This technique has lead to a self-consistent set of electron-HCl cross-sections shown in Figure 1. As indicated, the cross-section for excitation of the first vibrational level reaches a value in excess of 1.5×10^{-15} cm^2 slightly above threshold. Also shown in Figure 1 are the various contributions to the momentum transfer cross section, Q_m, the cross section for HCl dissociative attachment, and an effective electronic cross section which is most likely related to the direct dissociation process.

PRELIMINARY HCl CROSS SECTIONS



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The set of cross-sections presented in Fig. 1 can be used in conjunction with the calculated electron distribution function to determine the fractional power transfer to the various energy loss channels. Figure 2 presents the results of such a calculation as a function of E/n. Vibrational excitation of HCl is predicted to be quite efficient for $0.15 < \epsilon_K < 0.75$ eV.

A series of electron transmission experiments are underway within Professor George Schulz's laboratory at Yale University (Refs. 3 and 9). Some of these experiments have included an investigation of HCl in which the presence of a series of large resonances at low electron energies ($\epsilon_K < 5$ eV) has been detected. Ziesel and Schulz (Ref. 3) have been able to deduce cross-sections for excitation of HCl(v=1) and HCl(v=2) normalized to the absolute cross section for dissociative attachment. Using the dissociative attachment cross-section reported by Christophorou et.al. (Ref. 10), they calculate that the peak of the HCl(v=1) cross-section is about 1.5×10^{-15} cm^2, in excellent agreement with the value inferred by the UARL analysis of the Bailey and Duncanson transport data. Because the latter analysis also used the Christophorou cross section, the results of Yale experiments and the UARL calculations are consistent. Because the magnitude of the vibrational excitation cross section cannot be accounted for by direct excitation processes, the Yale group assumes that excitation proceeds by a resonant process involving $HCl^-(^2\Sigma^+)$.

VV and VT Relaxation Rates

The rate constant for the process

$$HCl(2) + HCl(0) \rightarrow 2HCl(1)$$

has been measured at room temperature by several investigators (Refs. 11-13), who report values of approximately 3×10^{-12} cm^3/sec. To scale this result to higher quantum numbers and different temperatures, the relative contributions of long- and short-range intermolecular forces must be known. It has been found that the magnitude of this rate at room temperature is accounted for by considering only short-range forces. The SSH expression as adapted for VV exchange by Keck and Carrier (Refs. 14, 15) is multiplied by factors of 1/2 and 1/9 to account for impact parameter and orientation averaging, respectively. Setting the range parameter L equal to 0.14Å gives agreement with the experimental value. Rates for higher quantum processes are obtained by applying Morse matrix elements. At low temperatures, this model predicts VV rates that are lower than would be the case if long-range forces made a contribution. Use of the short-range model is therefore probably conservative.

Zittel and Moore (Ref. 16) have used a laser fluorescence technique to measure the VT relaxation of HCl(1) by HCl and He in the temperature range 144-584°K. The self-deactivation rate has an inverse temperature dependence below room temperature, with $p\tau$ values, at 298 and 144°K, of 1.68 and 0.223 μsec-atm., respectively. Helium is 3 to 4 orders of magnitude less efficient as a deactivator than HCl itself, and therefore makes little contribution to the HCl relaxation.

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FRACTIONAL ELECTRON POWER TRANSFER IN HCl

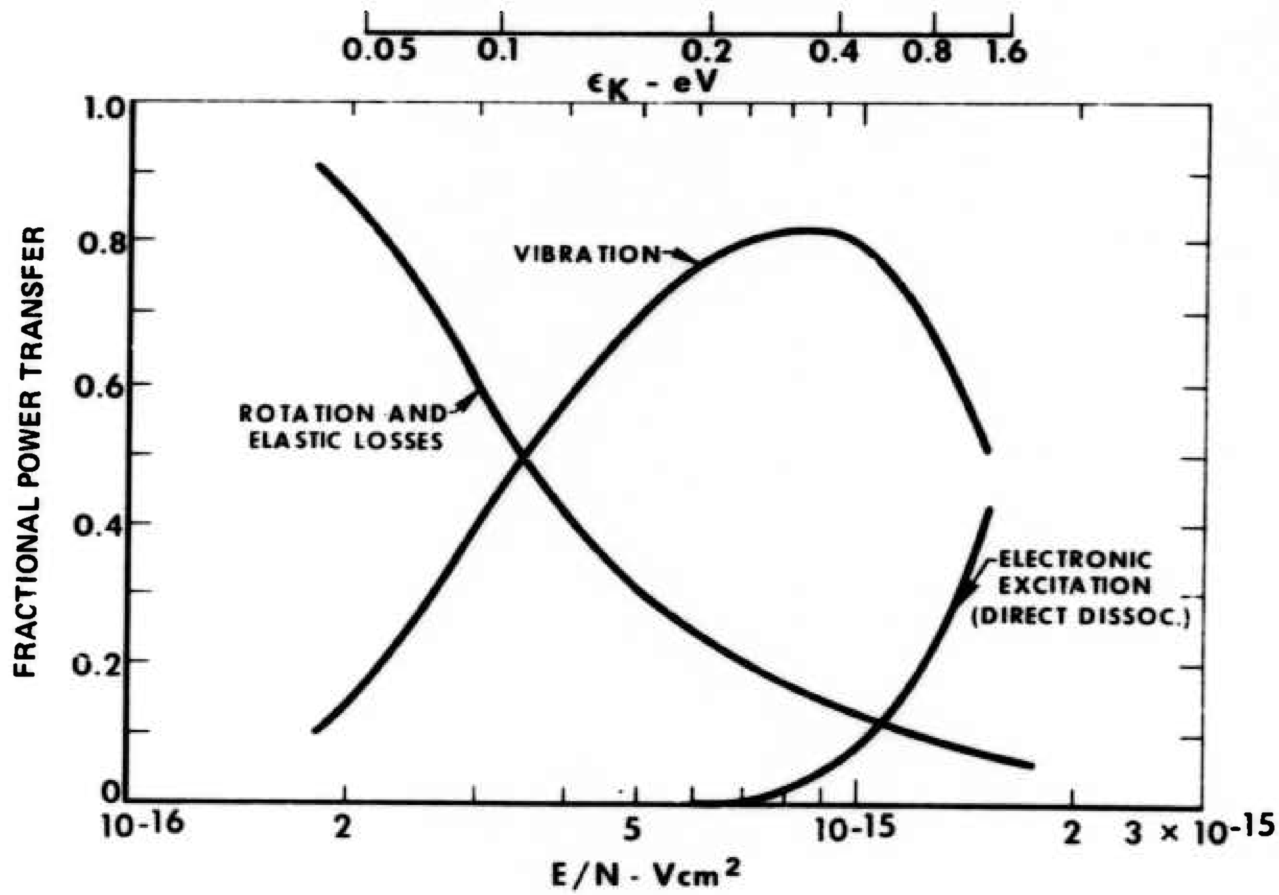

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FIG. 2

Optical Resonator Analysis

The optical configuration is assumed to be a transverse Fabry-Perot, or planar mirror system. Assuming geometrical optics, the solution to the radiative transfer equations reduces to solving the kinetic master equations for the plane-wave optical fluxes which maintain the loaded gains on oscillating transitions at the cavity threshold value. The P-branch having the maximum gain on any vibrational transition is assumed to oscillate preferentially.

Discharge Model

The conservative assumption is made that all discharge inefficiency (electrical power delivered to channels other than HCl vibration) results in heating of the translational-rotational degrees of freedom in the gaseous medium. Changes in bulk gas temperature and density are computed from ideal gas relationships. A constant sustainer potential is assumed in updating the Townsend parameter and hence the drift velocity and vibrational pumping rates.

Initial Calculated Results

The relationships governing the rates of change of the HCl state densities and bulk gas properties form a coupled set of first order differential equations. A computer code has been written which integrates these equations using the Euler-Cauchy technique (Ref. 18). A set of calculations have been performed to predict the time-varying characteristics of a representative HCl medium pumped by an electric discharge pulse at conditions presented in Table I.

Figure 3 presents the calculated vibrational distributions for several values of time after the onset of a 10 μsec exciting electric discharge pulse. Significant anharmonic pumping of higher vibrational states is predicted after 100 μsec. By 500 μsec, the distribution has relaxed considerably due to VT processes.

The profiles of small-signal gain coefficients corresponding to the computed distributions of Fig. 3 are shown in Fig. 4. While the $V=2 \rightarrow 1$ and $3 \rightarrow 2$ transitions show appreciable peak gain, the maximum gains for this set of conditions occur on the $5 \rightarrow 4$ and $6 \rightarrow 5$ transitions. Maximum gains of 3 - 4%/cm are predicted at $T \approx 150^\circ$K.

The desirablility of cooling the gas mixture to 150 - 200°K is shown in Fig. 5, where the computed gain profile on the $6 \rightarrow 5$ transitions is compared for T = 150 and T = 300°K. The computed peak gain increases by a factor of about 7 as a result of halving the translational-rotational temperature. Equilibration of rotational state populations with the translational temperature is assumed based on the rapid R-T relaxation measurements of Breazeale (Ref. 17).

Table I.

REPRESENTATIVE PULSED ELECTRIC DISCHARGE
HCl LASER MEDIUM CONDITIONS

Gas Mixture	
HCl	10.0 mol-%
He	90.0 mol-%
Initial Gas Properties	
Total pressure (const)	20 Torr
Translation temperature	150°K
Electric Discharge Pulse	
Pulse duration,	10 μsec
Electron density, n_e	$4 \cdot 10^{12}$ cm^{-3}
Townsend parameter, E/n_{HCl}	10^{-15} Vcm^2
Power deposition, P/n	500 W/cm^3
Optical Cavity	
Active optical path, L	56 cm
Mirror reflectivity, $r_1 = r_2$	99%
Mirror transmissivity, τ	0%
Threshold cavity gain	1.78×10^{-4}

CALCULATED POPULATION DISTRIBUTIONS IN PULSED HCℓ LASER

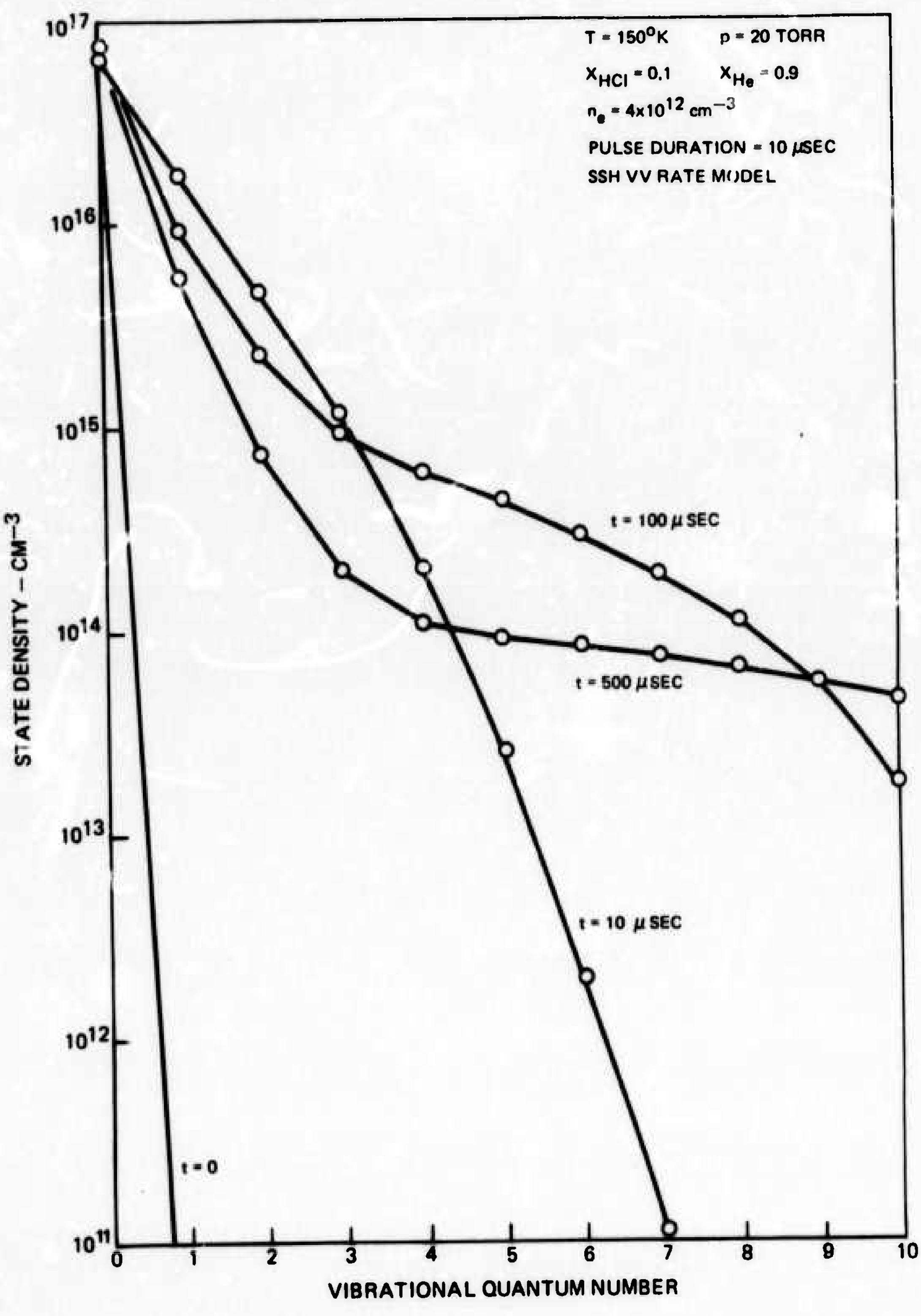

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FIG. 4

COMPUTED SMALL-SIGNAL GAIN COEFFICIENTS IN PULSED HCl LASER

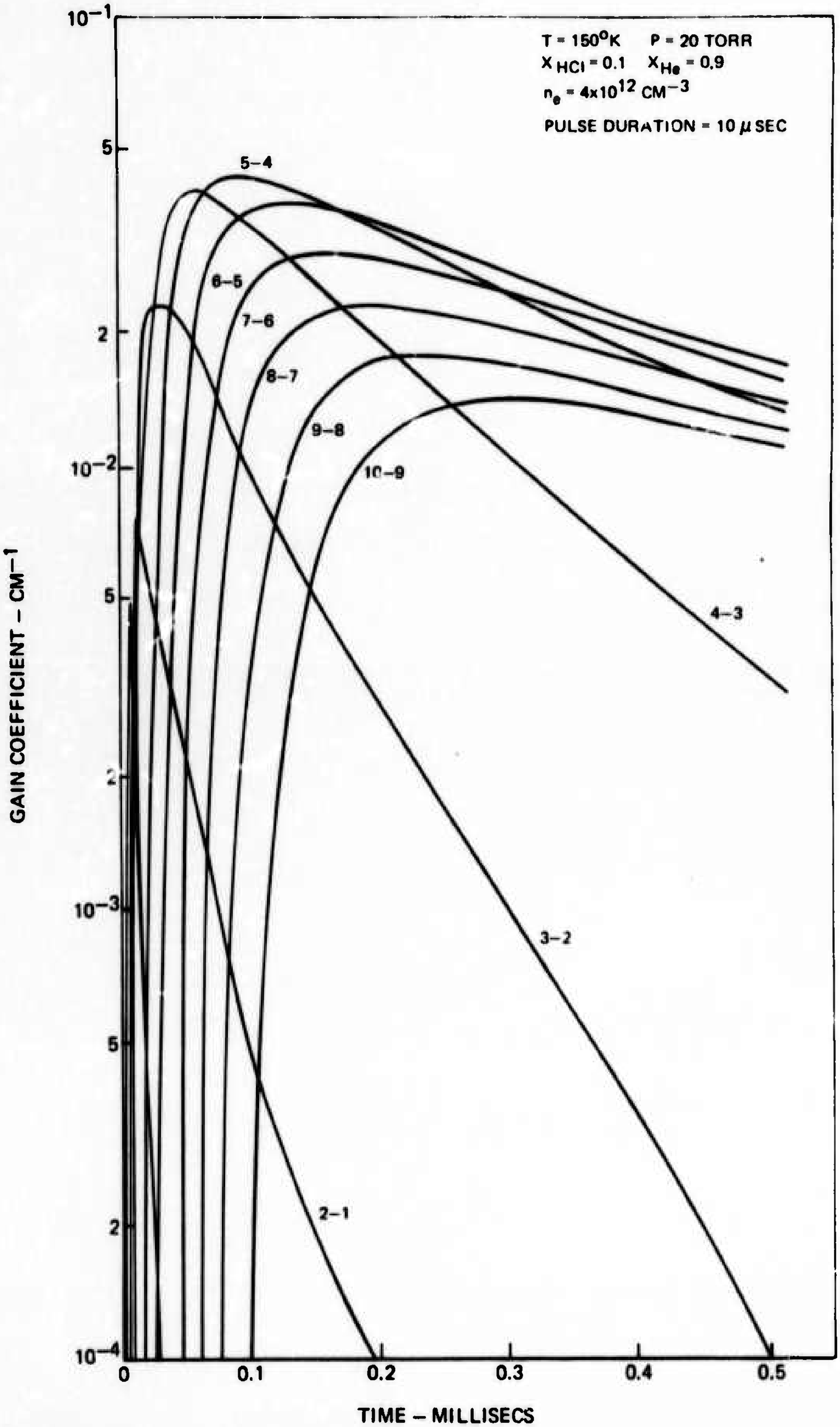


FIG. 5

VARIATION OF CALCULATED GAIN COEFFICIENT ON 6–5 TRANSITION WITH TEMPERATURE IN PULSED HCl LASER

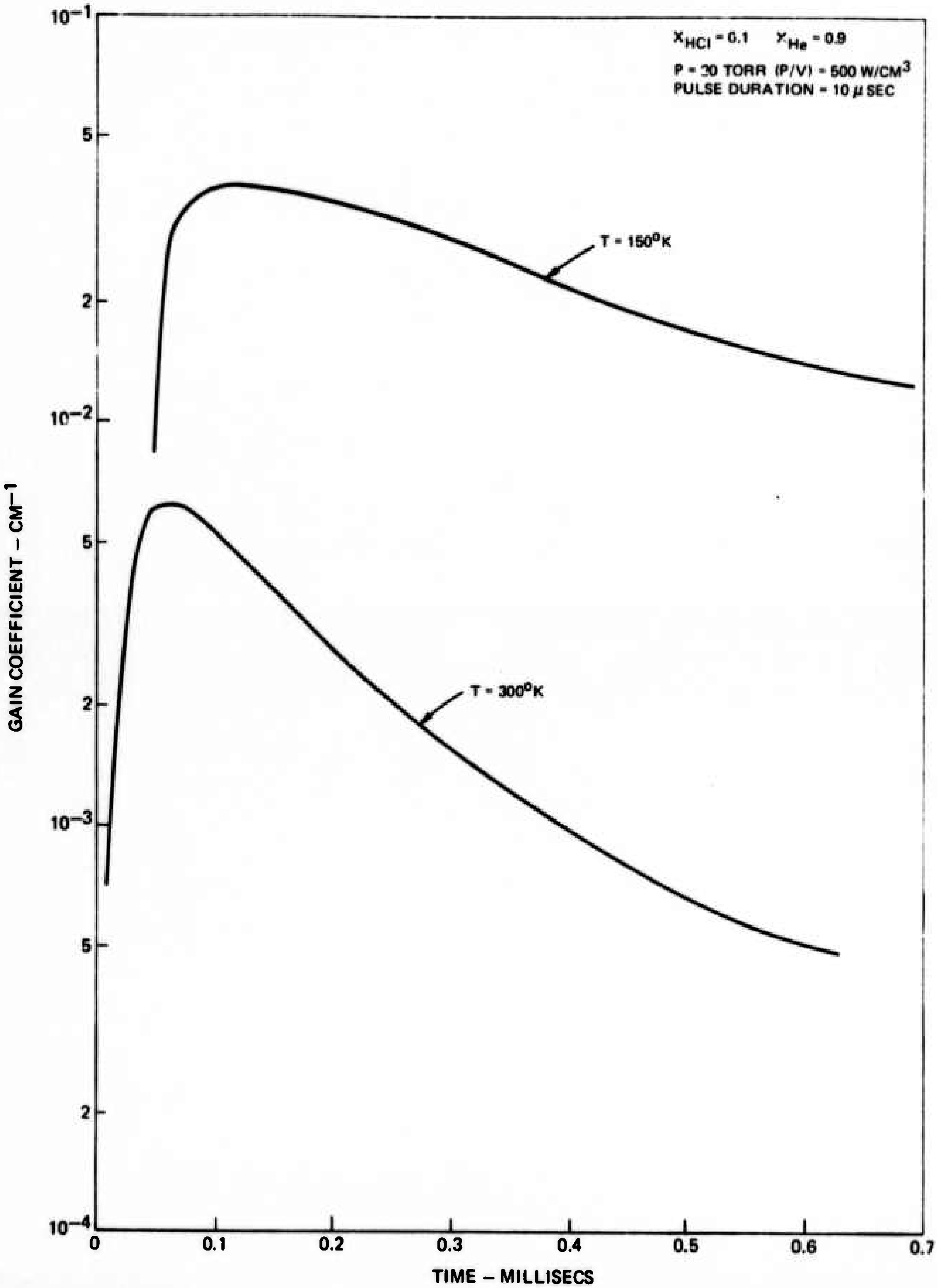
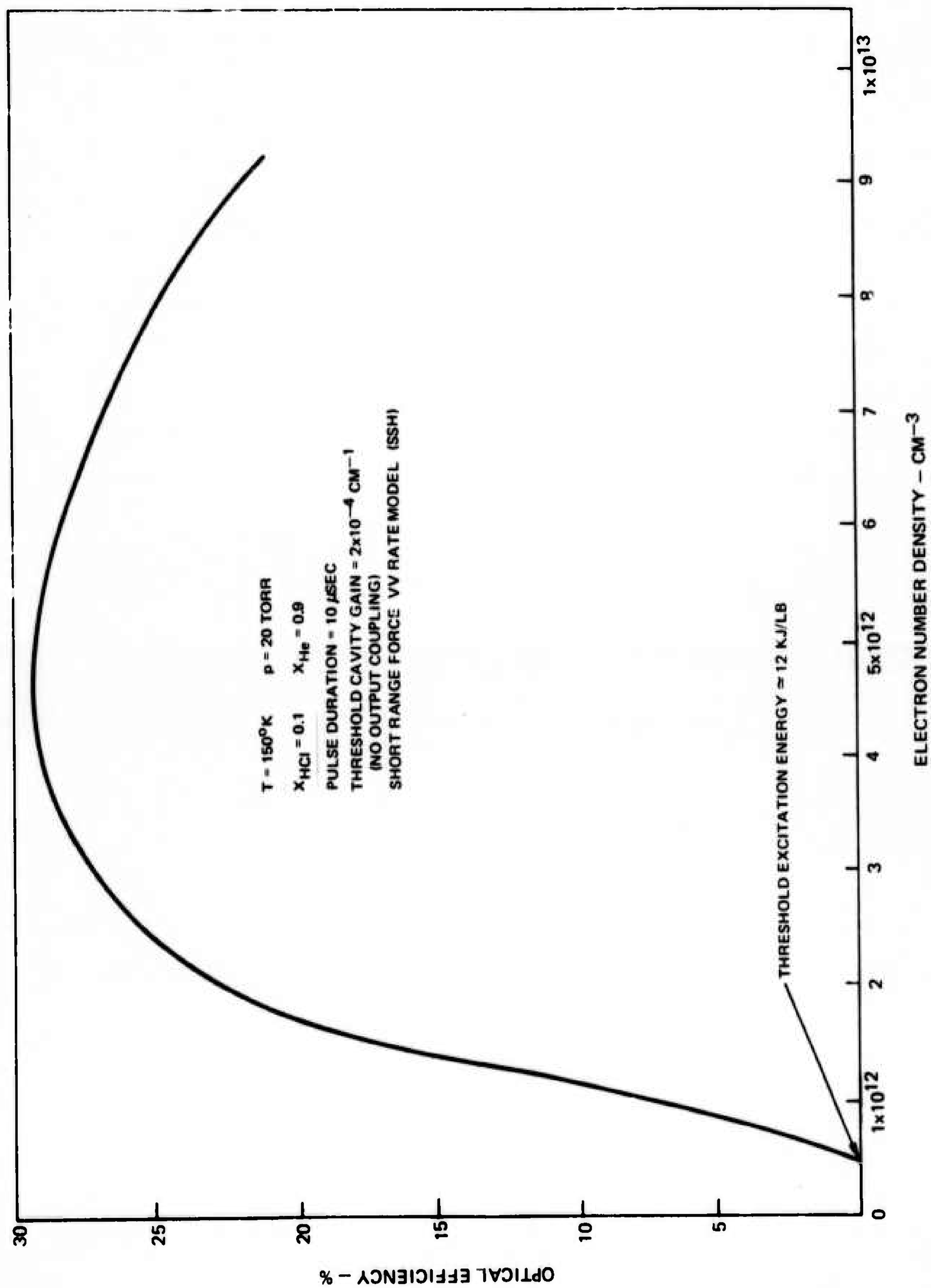


FIG. 6



COMPUTED OPTICAL EFFICIENCY VERSUS ELECTRON DENSITY FOR e–BEAM HCl LASER

Optical efficiencies have been calculated for a high-Q optical cavity (see Table 1). The variation of this quantity with initial electron density is shown in Fig. 6. A peak efficiency of about 30% is predicted. Assuming thet an optical extraction efficiency of 70% is attainable, overall output efficiencies of about 20% are indicated. At low values of n_e, the rate of energy deposition cannot compete with V-T relaxation. The zero optical power intercept yields a threshold excitation energy of about 12 KJ/lb for this set of conditions. For electron densities beyond a certain value, the rise of translational-rotational temperature due to discharge inefficiency becomes excessive, and the calculated efficiency decreases.

Experimental Progress

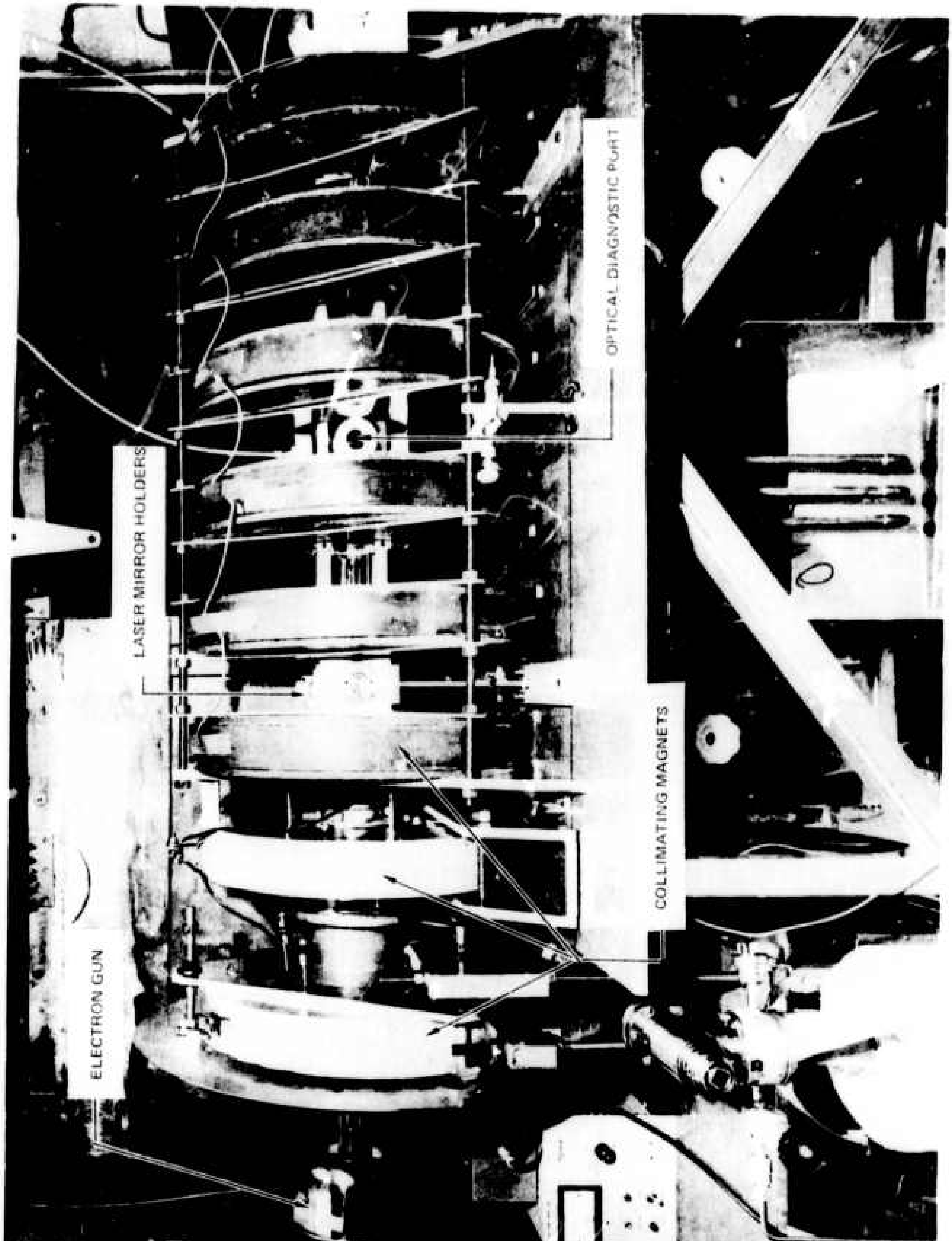
Experimental Apparatus

The experimental configuration conceived to demonstrate the emission of coherent radiation by electric discharge excitation is that of an electric discharge sustained by a high current density electron gun. The apparatus which has been assembled to demonstrate laser emission is shown in Fig. 7. An electron gun is fired down the axis of a discharge tube. A pulsed power supply provides a sustainer field synchronized to the high voltage pulse applied to the electron gun. The argon/hydrogen chloride gas mixture flows down the discharge tube toward the electron gun and exits from a sidearm near the gun. Gas flow rate is sufficient to complete an exchange of the discharge volume between pulses. Portions of the laser mirror holders as well as the optical diagnostic part may be seen in the photograph. The configuration of the laser may be best seen in Fig. 8. The laser resonator cavity will be desc ed later in detail.

The following section describes in detail various components of the experimental apparatus, as well as the experimental progress to date, here and at other laboratories.

The electron gun in use at UARL, has been described before (Ref.1,2). Briefly, the gun is composed of a barium oxide coated thermionic cathode and an anode with nineteen 0.250 in. diameter holes covered with an 0.8 mil aluminum foil to provide transmittal of the electron beam. Using this configuration and a seven stage Marx bank (charged to 20 kV per stage) with a 1000 ohm resistance in parallel with the gun, giving an RC time constant of 10 microseconds, to shape the high voltage pulse tail, a peak current density of 2 A/cm^2 has been observed with no foil degradation. It has been found that the gun performance and foil loading is dependent critically on the peak voltage of operation of this system. The exponentially falling voltage also presents a large heat load in the foil. This voltage waveform also presents primary electrons whose average energy is not steady in time. For these reasons it was decided to obtain an improved high voltage pulse power supply capable of applying reasonably square pulses. This unit will be described later in detail.

HCl PULSED ELECTRIC DISCHARGE EXPERIMENT



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In addition to the six magnet coils arranged along the discharge, two coils have been located in the region between the gun and the end of the discharge tube, as shown in Fig. 7. These coils are intended to maintain a uniform magnetic field into the gun cathode region and thereby avoid beam divergence associated with curving field lines at the ends of the magnet array. This array provides a reasonably uniform field of up to 1000 gauss on the discharge axis.

Optical Resonator

A stable resonator cavity has been designed and installed on the discharge tube in order to provide a long optical path length in the longitudinal geometry. Fig. 8 illustrates the geometry of the laser resonator. Mirrors internal to the vacuum chamber are set in adjustable mirror mounts held in sidearms blown in the discharge tube. Mirrors located opposite the end mirrors turn the optical axis so that the axis is skewed slightly with respect to the discharge tube centerline. The turning mirrors are held at the end of pedestals near the periphery of the discharge tube. The internal turning mirrors are 61 cm apart. The active optical path will be approximately 56 cm. The internal mirror/end mirror holder array is held stably by cer-vit rods separating the two holders. The vacuum seal to the glass is made with viton o-rings with a thin stainless steel diaphragm between the o-ring groove and the mirror holder. The two mirror arrays at either end of the tube are separated by additional cer-vit rods, so that the mirrors are held around the discharge tube independent of the thermal fluctuations of the discharge tube. Two sets of optics have been ordered and delivered. One set is a hole coupled gold coated optics and the other set is dielectric coated silicon with a partially transmitting output mirror.

New High Voltage Pulse Power Supply

The major component sub-assemblies of the newly installed, improved high voltage pulse power supply are shown schematically in Fig. 9. This pulser was designed and built by Systems, Science and Software (S^3) to UARL specifications. Basically the pulser is a three-stage Marx bank with a crowbar sparkgap to square off the tail of the high voltage pulse. The capacitance per stage ($0.05 \mu f$) is chosen so that the output voltage will not droop by more than 10 percent during a pulse. Rise and fall times are less than 1 μsec. The S^3 unit can produce 5-20μs duration pulses of 180 kV at current levels from 5 to 20 amperes, at a rate of 1 pps.

Considerable difficulty has been experienced in obtaining reliable operation of the HV pulser and extensive modfications have been made by the vendor since delivery. Upon delivery the output pulses fired into a dummy load appeared to be erratic in duration, becoming very erratic at a charge voltage of 60kV. The cause of this latter sympton was traced to the 35 ohm resistor in series with

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PULSED DISCHARGE LASER CONFIGURATION

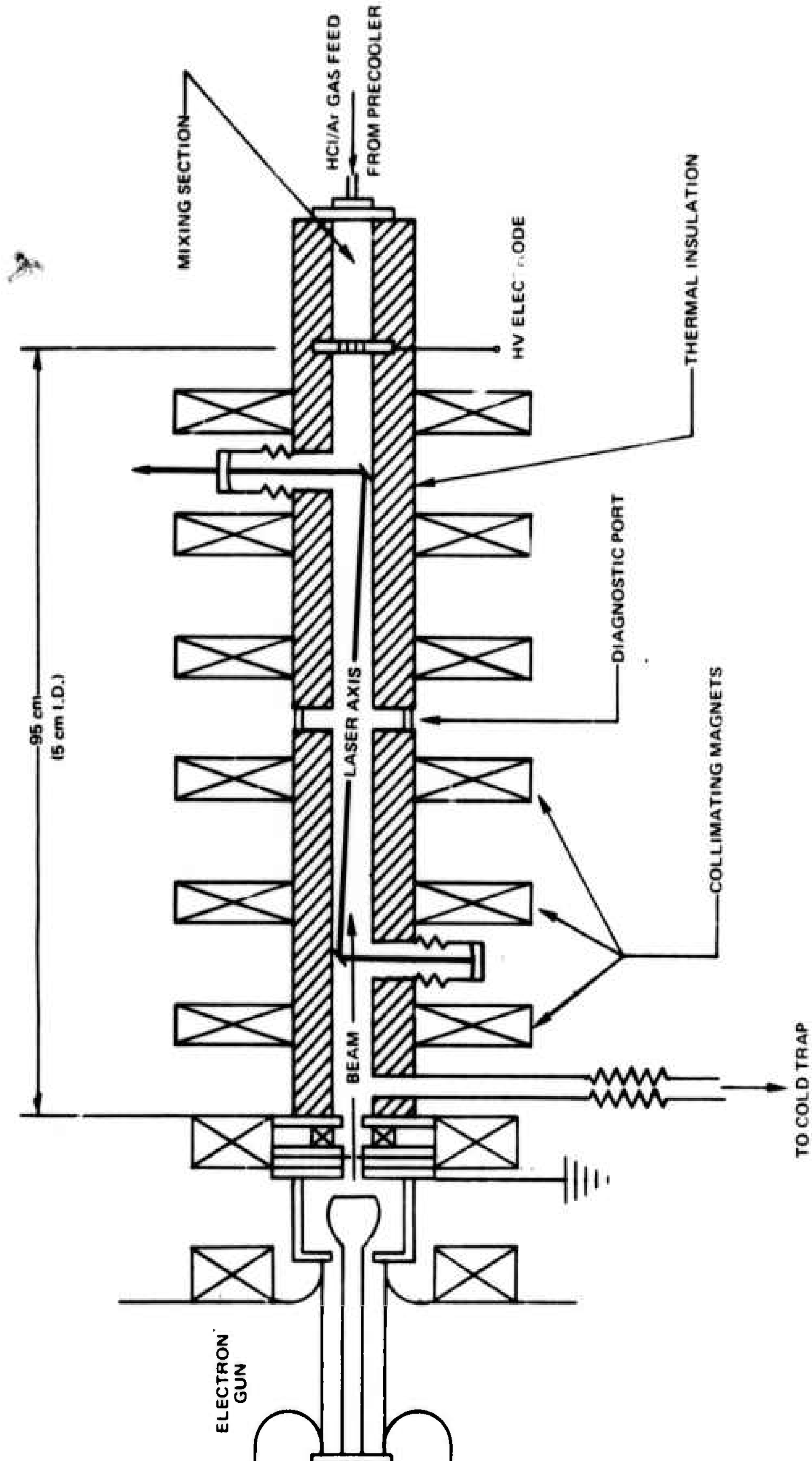
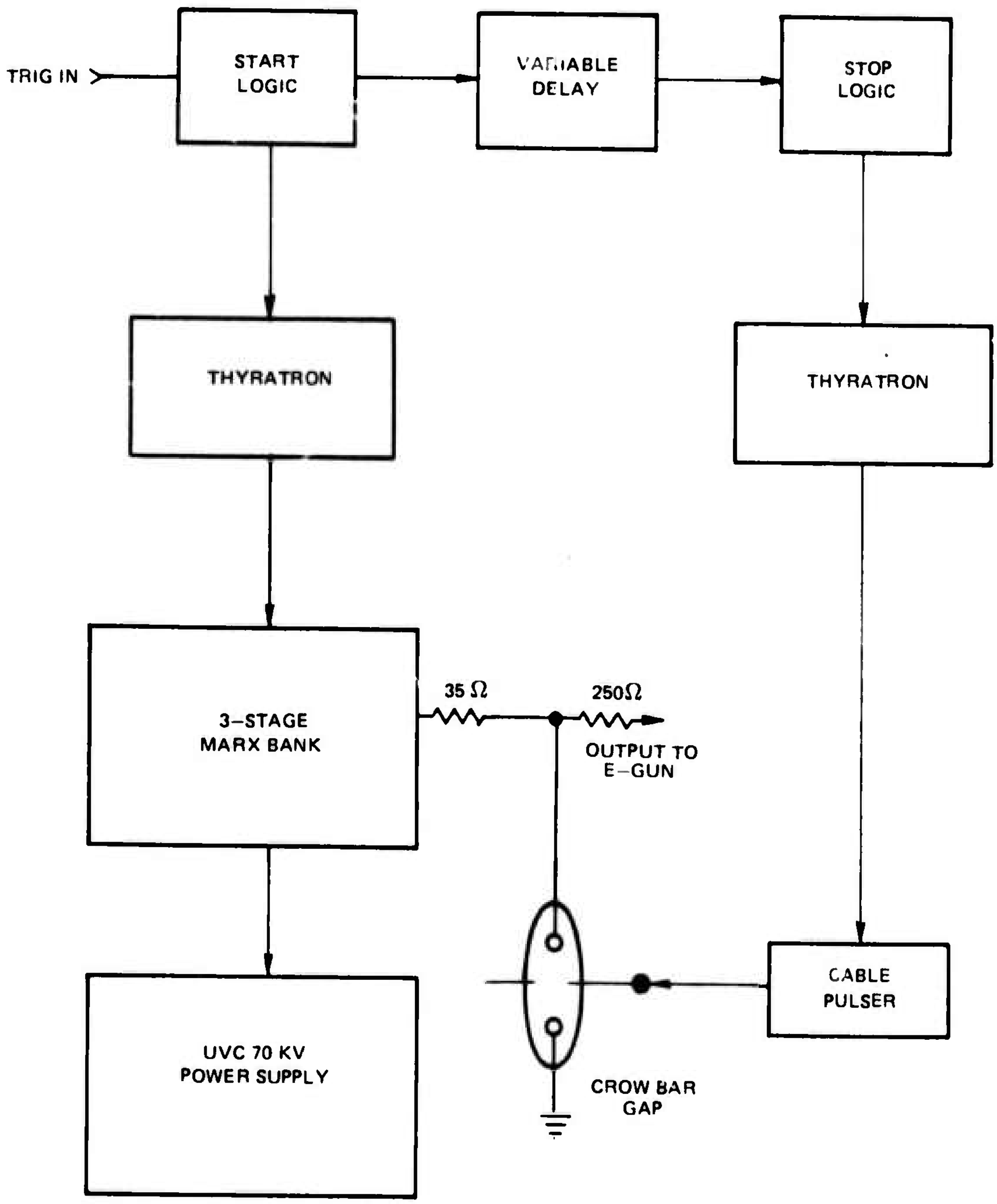


DIAGRAM OF SYSTEMS, SCIENCE AND SOFTWARE HIGH VOLTAGE PULSER



the output of the pulser. This resistor which is a stack of woven resistors was arcing between strands of the conductor. This component was replaced at the direction of the vendor with a stack designed for higher voltage and current rating, and which has operated satisfactorially. Irregular pulses of one of three kinds have been observed: (1) an essentially zero duration pulse, (2) a rapid pulse after a normal pulse and (3) a very long pulse of slowly falling voltage. This last kind of pulse would cause a failure of the electron gun through excessive heating of the foil. The vendor, S^3, decided to replace the crowbar circuit originally used with the arrangement shown in Fig. 9. The original crowbar circuit consisted of a conventional trigatron gap fired by a spark plug powered directly from the stop thyratron. The actual kind of erratic pulse observed depended on the operating pressure of the crowbar gap. Too little pressure resulted in a greater number of pulses of the (1) and (2) kind. Too great a pressure resulted in the (3) kind of pulse. The difficulty was there was no margin between these two pressure settings. That is, long pulses occurred too frequently to hope to apply to the electron gun. Upon a description of this behavior to the vendor, S^3 made some modifications to the low level logic and therafter decided to change the crowbar gap design to that shown in Fig. 9. The trigatron gap was replaced with a midplane gap triggered by a cable pulser consisting of a trigatron gap discharging a 20 ft. length of RG-17 coaxial cable charged by an independent 0-60 kV power supply. The trigatron trigger gap for the pulser was placed on an independent SF_6 gas supply. After these modifications were made, greatly improved performance was observed. The gas pressures in the three gap supply systems are still critical, however, there is a wider margin of operation so that erratic pulses occur less frequently. At present, difficulty with maintaining consistent operation at a fixed set of power supply operational parameters is being encountered. UARL is continuing to work closely with the vendor in overcoming these difficulties. The operational problems encountered with the S^3 high voltage pulse power supply precluded demonstration of laser action in HCl during this reporting period.

Experiments at Other Laboratories

Observation of Vibrational Excitation in HCl at Avco Everett Research Laboratories

In preliminary experiments undertaken by R. E. Center, et al., at AVCO Everett Research Laboratories (AERL) spontaneous emission from an HCl e-beam sustained discharge (Ref. 4) has been observed. To date stimulated emission has not been seen. By adding carbon monoxide to the mixture and monitoring relative spontaneous emission signals, AERL estimated that the vibrational excitation cross-section for HCl is comparable to CO. Thus, efficient vibrational excitation in HCl can be anticipated. Efficient vibrational excitation in HCl was predicted previously in the UARL theoretical study (Ref. 1).

Electric Discharge Laser Demonstration in HF and DF at Mathematical Sciences Northwest

Stimulated emission from low-lying vibrational transitions of HF and DF have been observed by S. R. Byron, et al., at Mathematical Sciences Northwest (MSNW) in preliminary, low energy output (~millijoule) experiments (Ref. 5). These results were obtained with an experimental configuration consisting of five electron guns ($J_b \simeq$ 50 mA/cm^2) firing transversely into a sustainer electric field. The active optical path length provided was 50 cm. Laser emission on the vibrational transitions $v=3 \rightarrow 2$, $2 \rightarrow 1$, and $1 \rightarrow 0$ have been observed in both HF and DF (multiple P-branch lines on certain transitions in DF). Laser action in HF has been seen both with and without H_2 present as an additive at approximately ten times the HF partial pressure. Argon and nitrogen buffers at 100 times the HF concentration have been effective. The total mixture static pressure was 190 torr and the discharge was at room temperature. Laser action in DF has been observed with Ar/DF mixtures at 99:1 molar ratios thus far. MSNW noted that the addition of hydrogen to the HF discharge produced better results. Whether this improvement is a result of additional vibrational energy transfer from H_2, or a shift in the chemical equilibrium of the mixture resulting in lower HF decomposition, is not known. A marked sensitivity of the output of the mixture electric field to neutral number density ratio was observed in accordance with previous UARL theoretical predictions (Ref. 1).

The experiments at AERL and MSNW both serve to confirm somewhat the behavior predicted by the UARL theoretical model reported previously (Ref. 1). The AERL experiment demonstrates that a substantial vibrational excitation rate can be obtained in an e-beam ionized, HCl electric discharge. The electron vibrational excitation cross section that they would derive from their data is approximately two to five times smaller than that predicted from early electron drift data (Ref. 1). The experiments at MSNW, although performed with HF (and DF), more or less confirm the UARL model predictions. Reference 1 suggested the addition of D_2 to the discharge to utilize vibrational energy transfer from electron pumped deuterium vibrational states. The MSNW experiments demonstrated high gain on low-lying vibrational transition with low partial pressures of active species as predicted. Furthermore these preliminary experiments illustrate the necessity for careful E/n control to operate in a regime of high vibration excitation with minimal rotational and electronic excitational losses. Lastly the requirement for large electron densities in these pulsed dicharge lasers is confirmed. In summary, these other preliminary experiments illustrate that hydrogen chloride can be excited effectively in a pulsed, externally ionized discharge, and should be capable of lasing, given the proper conditions.

Future Studies

Future investigations of the possible HCl electric discharge laser are planned to include (1) demonstration of stimulated emission, (2) optimization of stimulated emission output energy and (3) characterization of the lasing medium and description of the generated output.

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